

Angle-Resolved, Two-Dimensional Imaging Reveals Significant Effects

Although the details of the fine structure of multi-electron systems cannot be calculated exactly (because of perturbations caused by interactions between the electrons), the LS coupling approximation is widely assumed to be valid, at least for low-lying excitations in light atoms. However, researchers at the Advanced Light Source (ALS), using angle-resolved, two-dimensional photoelectron spectroscopy, have found that LS-coupling predictions are not only violated, but that the effects of electron-electron interactions are unexpectedly large and cannot be ignored when studying detailed spectra in atoms as light as neon.

Doubly excited systems, where one photon excites two electrons, clearly demonstrate electron-electron interactions, since one photon can excite only one electron directly. Thus, studies of doubly excited systems using high-precision instrumentation and high-

resolution photon sources can reveal details about atomic structure that result from electron-electron interactions. Such studies have been conducted successfully on simple atoms such as helium and lithium. Photoabsorption results from early experiments on a more complex system, neon, have demonstrated a departure from the predictions of LS coupling in the fine-structure splittings of the photoionization spectra. While these results established the existence of certain electron-electron interaction effects, they lacked additional information that would be provided by angle- and energy-resolved measurements. Furthermore, only *small* electron-electron effects were observed.

The ALS researchers performed more complex, detailed studies of the photoexcitation and decay mechanisms of doubly excited neon in order to probe electron-electron interaction effects in this relatively

light system. Neon is more complicated than helium and lithium because of its occupied 2p subshell. In doubly excited, photoionized neon, one photoelectron is emitted and one electron is boosted to a higher energy level, leaving the ion in one of several available energy states. The dense grouping of these states results in photoionization spectra with a complex structure. These states reveal higher-order electron-electron interaction effects and provide a stringent testing ground for the accuracy of existing computational methods.

Two-dimensional photoelectron spectra (electron yield as a function of photon energy and binding energy) were taken at two angles (0° and 54.7°) simultaneously, in order to efficiently observe any effects or features. The apparatus consisted of two highly efficient time-of-flight electron-energy analyzers. The measurements were performed on Beamline 9.0.1, an undulator beamline

for high-resolution atomic and molecular studies.

Several features of the resulting spectra indicate violations of LS-coupling predictions. For example, in this study, according to LS coupling, signals should vanish at 0° , so that the observation of any signal at this angle is an immediate indication of the breakdown of LS coupling. The data, which could only have been obtained through the ALS's capability to provide high-resolution photons coupled with the capability to view the data in two dimensions, revealed unexpectedly large effects due to electron-electron interactions. These results have been corroborated by state-of-the-art *ab-initio* calculations. The important implication of the detection of prominent electron-electron interaction effects is that it is not safe to assume the validity of LS coupling, even for low-lying excitations in a system as light as neon.

N. Berrah (616-387-4955/510-495-2439), Department of Physics, Western Michigan University.

A. A. Wills, T. W. Gorczyca, N. Berrah, B. Langer, Z. Felfli, E. Kukk, J. D. Bozek, O. Nayandin, and M. Alshehri, "Importance of Spin-Orbit Effects in Parity-Unfavored Photoionization of Neon, Observed Using a Two-Dimensional Photoelectron Imaging Technique," *Phys. Rev. Lett.* **80** (1998) 5085.



ELECTRON-ELECTRON INTERACTIONS IN NEON

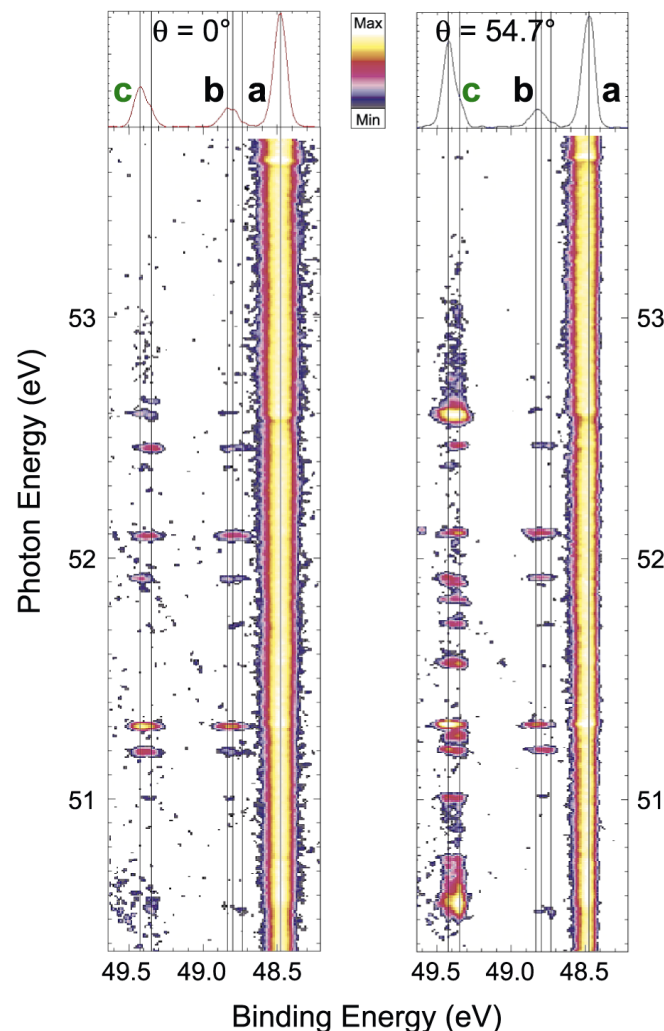


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- **Calculation of atomic energy levels**
 - *Exact solutions precluded by interactions between electrons*
 - *LS-coupling approximation widely used*
- **Study of doubly-excited neon**
 - *Probe higher-order electron-electron interaction effects*
 - *Stringent test of theoretical models*
- **ALS facility and state-of-the-art instrumentation allow**
 - *High-resolution data*
 - *Angle-resolved data (obtained at 0 degrees and 54.7 degrees)*
 - *Two-dimensional data (photon energy vs. binding energy)*
- **Results show**
 - *Violations of LS-coupling predictions*
 - *Larger-than-expected electron-electron interaction effects*
 - *Importance of electron-electron interactions when studying detailed spectra in atoms as light as neon*

ELECTRON-ELECTRON INTERACTIONS IN NEON

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Photoelectron yield as a function of photon energy and binding energy at 0 degrees and 54.7 degrees.

The upper graphs show the spectrum at a photon energy of 51.3 eV, and the vertical lines a, b, and c indicate the positions of various fine-structure energy levels.

The signals along vertical lines c at 0 degrees indicate the breakdown of LS coupling.